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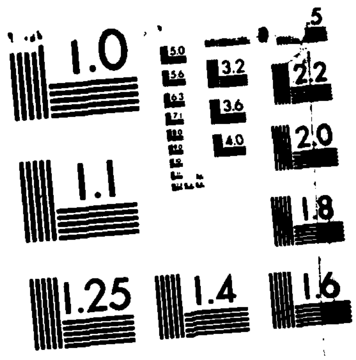

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INVESTIGATION ON GROWTH AND LASER PROPERTIES OF GGG: (Nd,Cr) SINGLE CRYSTALS

by

Zhang Lehui, Lin Chengtian, et al.

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INVESTIGATION ON GROWTH AND LASER PROPERTIES OF GGG:(Nd,Cr) SINGLE CRYSTALS

by

Zhang Lehui, Lin Chengtian, Liu Hairun, Liu Lin,
Zhu Huanan, and Lin Xi
Institute of Physics, Academia Sinica

ABSTRACT Investigation on the growth and laser properties of gadolinium gallium garnet crystal doped with neodymium ~~Nd~~ and chromium ~~Cr~~ is reported. As the segregation coefficient of Nd in GGG is less than 1 and that of Cr is greater than 1, a modified Czochralski method for growth is adopted in order to keep the dopants being uniform in the grown crystal. (Translations, China, Chinese language)

1. INTRODUCTION

Gadolinium gallium garnet doped with Nd (GGG:Nd) has been widely reported as a laser operating material.[1-5] Since the segregation coefficient of Nd in GGG:Nd is larger than that in YAG:Nd during the single crystal growth (0.5 for GGG:Nd[3,4] and 0,21 for YAG:Nd), it is more feasible to dope GGG with Nd. This fact has interested those who perform crystal growth and those who use the laser rods. However, GGG:Nd crystals produce color centers easily after an exposure to the UV radiation. This is their disadvantage as a laser operating material. This paper discusses the crystal growth and properties of the Nd and Cr doped GGG. Since the Nd

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dopant increases the lattice constant, we tried to reduce lattice aberration by selecting another ion as a dopant to reduce the lattice constant. This is the reason why we used the double-dopants. In an attempt to find GGG crystals capable of resisting radiation, we measured the spectra for the double-dopant GGG:(Nd, Cr) crystals, their laser output and the color center absorption after irradiation.

2. EXPERIMENTS

2.1. Double-dopant Selection

Before the crystal growth, it is essential to investigate the properties of the double-dopant ions. Doping with Nd makes GGG lasing, but increases in its lattice constant.[3] Hence, the principle of the double-dopant selection is to find another ion and simultaneously dope the GGG crystals with the two ions to reduce the lattice constant without lattice aberration. After comparing various ions of different radii,[6] Cr^{3+} ion was chosen as the second dopant. The GGG crystals doped with Cr were tested and its lattice constant measured. Table 1 lists the variations of the lattice constant after GGG single crystals were doped with either Nd or Cr. The Cr^{3+} ion enters the center position of an octahedron in the lattice, reducing the lattice constant of the GGG crystals and probably compensating for the lattice aberration resulting from the substitution of larger Nd^{3+} for Gd^{3+} (entering the center of the dodecahedron). In addition, the energy levels of Cr^{3+} and Nd^{3+} in the garnet crystals are overlapping, and energy transfer can possibly occur. Since this sensitization would be beneficial to lasing, the two ions, Nd^{3+} and Cr^{3+} were chosen as the dopants and

Table 1. Changes in the lattice constant of a GGG single crystal after Nd or Cr doping.

1-晶 体	2 掺Nd浓度* 3 (原子百分比)	4 掺Cr浓度** 5 (重量百分比)	晶格常数*** 6 (Å)
GGG(Nd)	0.5		12.381
	1.5		12.383
	2.5		12.385
	3.5		12.386
GGG(Cr)		0.045	12.3814
		0.14	12.3804
		0.18	12.3800
		0.30	12.3792
		0.33	17.3740

1-crystal; 2-Nd concentration; 3-at%; 4-Cr concentration; 5-wt%; 6-lattice constant.

Note: *literature [3]. **Liu Ling et al., An Investigation on Crystal Growth and Color of the Doped GGG, to be published. ***The range of the lattice constant of the pure GGG single crystals we measured is 12.382±0.00Å.

the double-dopant GGG crystals were grown.

2.2. Measurements of Crystal Segregation Coefficient and Optical Transmittance, and Selection of the Growth Technique

Since, during the GGG:Nd single crystal growth, the segregation coefficient of Nd in GGG is less than 1, the concentration of Nd in the crystal growth by using the common pulling techniques gradually increases with the length of the crystal. In order to measure the segregation coefficient of Cr in GGG during crystallization, we also grew the Cr-doped single crystals. Since the segregation coefficient of Cr is 2.7, concentration of Cr decreases considerably as the length of the rod increases during pulling. The transmittance curves for GGG:Nd, GGG:Cr and GGG:(Nd,Cr) single crystals are shown in Figs. 1

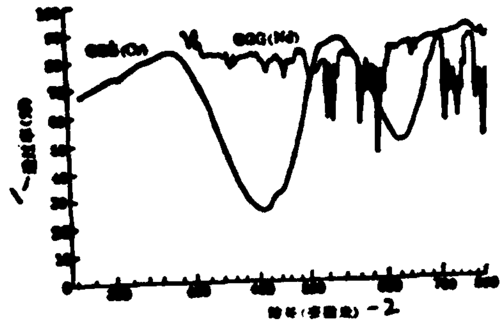

Fig. 1. Optical transmittance curves for GGG:Nd and GGG:Cr single crystals.

1-transmittance; and 2-wave length (nm).

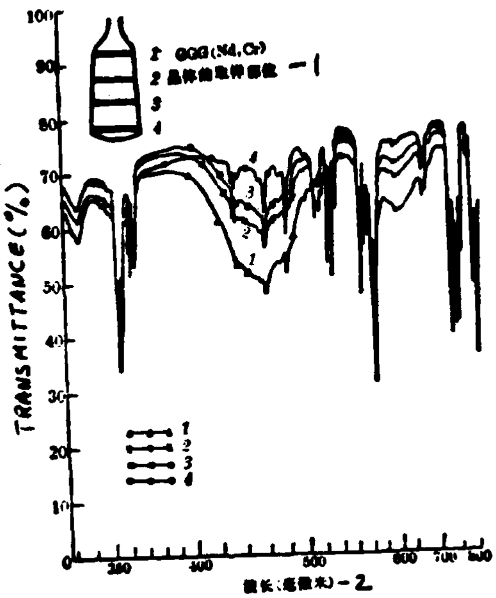

Fig. 2. Optical transmittance curves for the slices taken from four different positions on the GGG:(Nd,Cr) single crystal grown using the common pulling techniques.

1-sampling positions on the GGG:(Nd,Cr) crystal; 2-wave length (nm).

and 2.

In order to ensure the uniformity of the dopants in the GGG:(Nd,Cr) laser rods, a modified Czochralski method was adopted. This method permits one to keep the crosswise variations of the Nd and Cr concentrations small, and to ensure the homogeneity of the dopants within the whole rod.

2.3. Measurement of the Color Center Absorption Spectra for the Single Crystals

The four grown crystals, GGG:Nd, GGG:Cr, GGG:(Nd,Cr) and GGG without dopant were cut into slices which were 1-4mm thick. Duplicate samples were prepared from each kind of crystal with both polished sides, one sample being exposed to the γ-radiation (a $\sim 10^6$ roentgen dose) using 30MeV linear accelerator and the other not. The two sets of samples, irradiated and unirradiated, were compared and the color center absorption spectra were run for the irradiated crystals, as shown in Fig. 3.

Long slices were also cut from the crystals lengthwise. They were exposed to the UV radiation (30w for 1 hr), and their transmittance curves were run on various positions of these slices. After comparing these curves with those for the corresponding unirradiated slices, it was found that irradiation has small influence on transmittance for Nd-doped GGG crystal slices containing a certain amount of Cr.

2.4. Measurements of the fluorescence spectra in the visible region and the fluorescence lifetime for GGG:(Nd,Cr) crystals

The fluorescence spectra in the visible region were measured

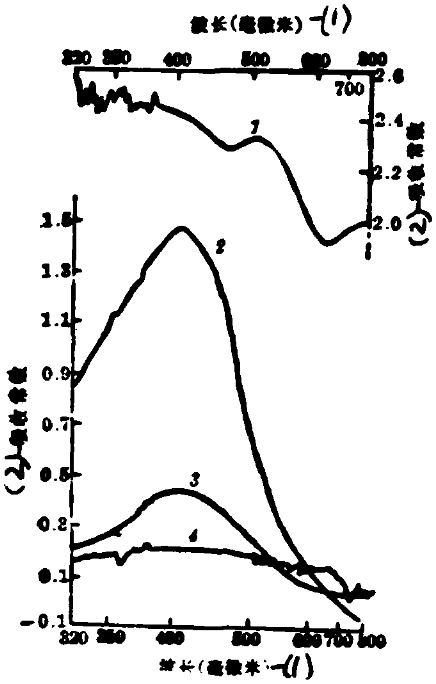

Fig. 3. The color center absorption curves for the doped GGG crystals.

1-GGG:Cr, irradiated with 10^5 roentgen;

2-GGG:Nd, irradiated with 10^5 roentgen;

3-pure GGG, irradiated with 4.3×10^6 roentgen;

4-GGG:(Nd,Cr), irradiated with 10^5 roentgen.

(1)-wavelength (nm); (2)-absorption coefficient.

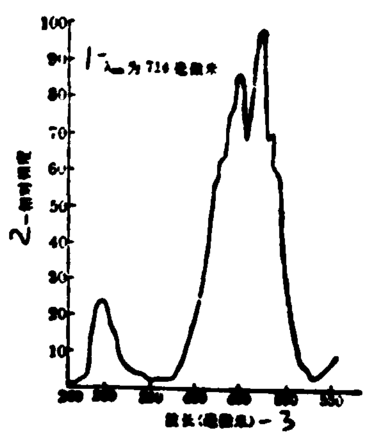

Fig. 4. The excitation spectrum of GGG:(Nd,Cr) single crystal, λ_{em}=714nm

1-λ_{em}=714nm;

2-relative intensity;

3-wavelength (nm).

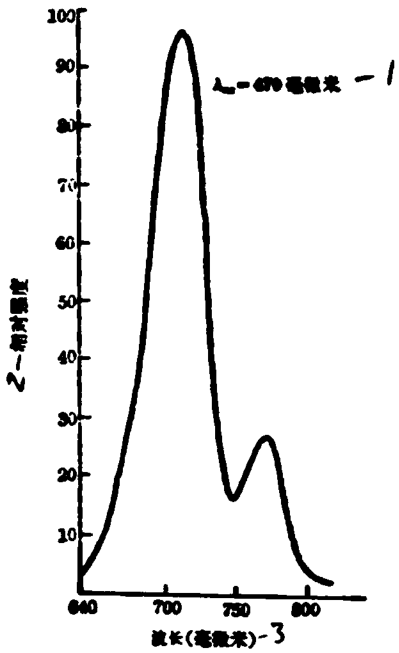

Fig. 5. The fluorescence spectrum of GGG:(Nd,Cr) single crystal, λ_{ex}=470nm.
1-λ_{ex}=470nm;
2-relative intensity;
3-wavelength (nm).

for the GGG:(Nd,Cr) single crystal slices. The excitation and fluoresence spectra are shown in Figs. 4 and 5. Fig. 4 is the excitation spectrum with 714nm radiation of Cr^{3+} ions in the crystal. The two bands at 470 and 625nm correspond to $4A_2 \rightarrow 4T_1$ and $4A_2 \rightarrow 4T_2$ jumps and the spectral widths are 600~1000Å. Fig. 5 is the fluorescence spectrum of Cr^{3+} with 470nm excitation.

The following are the results of the fluorescence band lifetime for Nd^{3+} and Cr^{3+} in the GGG:(Nd,Cr) single crystals:*

Nd^{3+} $4F_{3/2} \rightarrow$ peak value is 1.06μm, T_f=250μs.

Cr^{3+} $4T_2 \rightarrow$ peak value is 714nm, T_f=105μs.

where T_f is the fluorescence lifetime.

2.5. Laser Output Measurement

A 5mm in diam. by 30mm long rod was prepared from GGG:(Nd,Cr) crystal. The edge surfaces of the rod are not coated and are

*Data provided by Wu Guangzhao, Chen Shuchun and Zhang Xiurong of the Shanghai Institute of Optics and Fine Mechanics, Academia, Sinica.

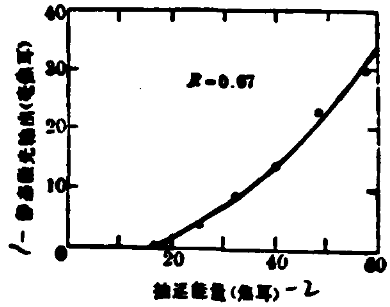

Fig. 6. Laser output energy of the GGG:(Nd,Cr) rod.

1-static laser output (mJ);

2-extracted energy (J).

perpendicular to the rod axis. The rod was positioned in a single ellipse cavity with silver film linings (originally this was a condenser where a 6mm in diam. by 65mm long YAG:Nd rod was placed), and optical pumping was conducted using a 7mm in diam. by 70mm long xenon tube lamp. Between the rod and the lamp there was a UV glass filter. The whole cavity was cooled by sodium nitrite solution.

The capacitance to store the energy for the xenon lamp to discharge is 80μF. The glow of the lamp discharge lasts 150μs.

The cavity is a plate-plate chamber structure where one mirror has complete reflection at 1.06μm and the other only partially reflects. The cavity is 0.54m long. Fig. 6 shows the variation of the static laser output with energy extraction when the reflection index of the output mirror equals 67%.

3. DISCUSSION

3.1. Measurement of the Absorption Spectra for the Single Crystals

It is shown from Fig. 2 that the transmittance curves for the GGG:(Nd,Cr) single crystals are basically an addition of those for the two crystals, GGG:Nd and GGG:Cr. The variations of the Nd^{3+}

and Cr^{3+} absorption peak heights indicate the difference in the Nd and Cr concentrations at positions 1, 2, 3 and 4 of the crystal after pulling. It can be seen that in the grown crystals using the common pulling techniques, the dopant concentration in a laser rod is inhomogeneous lengthwise. The absorption spectrum around 455nm can be taken as the characteristic spectrum of Nd^{3+} and Cr^{3+} where the absorption due to Nd^{3+} appears as two sharp peaks, and that due to Cr^{3+} as a broad band. From position 1 to position 4 of the crystal, the Cr^{3+} absorption band became weaker and weaker, while the Nd^{3+} sharp peak gradually increased. These changes in the intensities of the absorption spectra indicate that the dopants in the crystal rod grown by the common pulling techniques are very inhomogeneous. When cutting the crystal crosswise at certain levels, the Nd and Cr concentrations in the rods can be kept generally unchanged.

3.2. Single Crystal Doping, Color Center Absorption Peak Intensity and Behavior of the Irradiation Resistance

The color center absorption curves for the γ-ray irradiated GGG:Nd, GGG:Cr and GGG:(Nd,Cr) crystals are shown in Fig. 3. It is evident that the color center absorption peak has a lower intensity for the GGG crystal with double dopants, Nd and Cr than with single dopant, Nd or Cr. This indicates that the former crystal is more irradiation-resistant than the latter. The fact that the color center concentration in the GGG crystals with double-dopants is smaller than that with single dopant might be explained in terms of small lattice aberration. As mentioned above, after doped with Nd, the GGG lattice was enlarged while doping with Cr made the lattice small, and the double-dopants in the GGG can reduce the lattice

aberration. This probably is the reason why the color center concentration in GGG:(Nd,Cr) is small after irradiation. In addition to exposure to the γ-radiation, the same effect was observed under UV radiation.

3.3. Measurements of the Gain Factor and Loss, and Comparison of these properties with YAG:Nd

The 5mm in diam. by 30mm long GGG:(Nd,Cr) rod and 6mm in diam. by 65mm longYAG:Nd rod were successively placed in the laser cavity mentioned earlier and the laser oscillation threshold values with various reflection indexes of the output mirror were measured, from which the gain and loss of the laser operating materials can be obtained.[8] Fig. 7 shows the variation of small signal single-pass gain measured in the laboratory for these two laser operating materials with the pumping power (pumping energy per unit volume of the operating material). It can be seen from the figure that with the same pumping power, the gain factor of GGG is about a half of

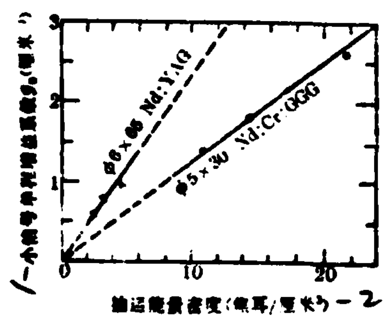

Fig. 7. Variation of the gain factor with the pumping energy density for GGG:(Nd,Cr) and YAG:Nd.
1-small signal single-pass gain factor, g_0 (cm^{-1});
2-pumping energy density (J/cm^3).

that of YAG. The same experiment also shows that the losses for the GGG and YAG rods are 0.68 and 0.13, respectively. It is believed that as the techniques for GGG crystal growth with double-dopants are improved, the loss will decrease.

It should be mentioned that as a laser operation material, GGG:(Nd,Cr) still has a long way to go, when compared with YAG:Nd. Nevertheless, the advantages of the former material show that it is worth further investigating.

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